TITLE: ULTRAVIOLET PHASE CONJUGATION AND ITS PRACTICAL
IMPLICATIONS

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Abstract

We report the first demonstration of uv phase conjugation. Using a 15 psec, 2660 Å pulse, 0.1% conjugate reflectivities were obtained via degenerate four-wave mixing in 1-mm samples of CS_2 mixtures. While pure CS_2 did not exhibit the effect, dilution in several uv transmitting solvents opened up a concentration-tunable (2450 Å - 2850 Å) spectral window, allowing the optical Kerr effect to be utilized. Weaker phase conjugation at 2660 Å was also observed in other Kerr media and in saturable absorber media.

Introduction

The desirability of phase conjugation in the uv is widely acknowledged. A natural choice for an efficient conjugator would be the highly nonlinear liquid CS_2. Unfortunately, pure CS_2 is opaque over most of the uv spectrum. However, we have found that by diluting CS_2 in uv-transmitting solvents, the absorption bands are significantly shifted, opening up a concentration-tunable transmission window in the important 2450 Å to 2850 Å range. Establishing this transmission window in CS_2 allows one to take advantage of its large Kerr effect, which, in the uv, is enhanced over its visible value.[1] Within this concentration-tunable window, we have obtained a phase-conjugate 2660 Å, 0.1% reflection in a 1 mm mixture (by volume 60% CS_2, 40% hexane), consistent with the large nonlinearity of CS_2.

In degenerate four-wave mixing,[2] a probe wave impinges upon a nonlinear material that is illuminated by a pair of counterpropagating pump waves. The medium reradiates a fourth phase conjugate wave precisely retracing the k-vector of the probe wave even though an aberrator may be in the way.

These distortion-correcting features of phase conjugation suggest widespread applications, in particular in the uv. First, all high-energy uv lasers are high-gain excimer systems, characterized by severe optical inhomogeneities. These lasers are prime candidates for laser fusion, laser isotope separation, and very long distance propagation. An appropriate conjugator could minimize the influence of static aberrations both in the laser and between the laser and the target.[3] A highly aberrated excimer laser system could thus be converted into one providing a well-collimated, highly focusable beam. As a second application, uv phase conjugation could be used in conjunction with photolithography (or other laser processing techniques) to produce ultraminiature integrated circuit components. Lastly, in the uv, optical tolerances are hard to attain; phase conjugation may permit inferior elements in uv optical systems.

Solvatochromism in CS_2

In choosing nonlinear uv phase conjugation samples, we sought transparent materials with a strong optical Kerr effect and a short response time. Although liquid CS_2 is useful for visible and ir wavelengths, it is opaque below 3600 Å because of strong and broad electronic absorption bands at 3100 Å and 2200 Å. Nevertheless, we have observed that when CS_2 is mixed with uv-transmissive organic solvents, a window appears between the two absorption bands whose width and center frequency depend upon the specific solvent and its concentration.

Figure 1 shows uv transmission curves for 1-mm-thick samples of various mixtures of reagent grade CS_2 and uv grade hexane. The region of transparency broadens and the peak transmission wavelength blue shifts by 3000 cm^{-1} as the CS_2 concentration is reduced. Without solvent-solute interaction, the peak transmission wavelength would not have changed. Although solvent shifts for dilute systems are well-known,[4] we are not aware of any concentration-dependent studies in CS_2-hexane. At 60% CS_2 concentration, the 2660 Å transmission is 50%, making this mix suitable for phase conjugation at this wavelength and at the 2680 Å wavelength of CH_4-Stokes' shifted KrF laser light. Further dilution of the CS_2 increases the transmission at both 2485 Å and 2820 Å, suggesting that longer dilute samples would be suitable phase conjugators at KrF and XeBr excimer wavelengths.
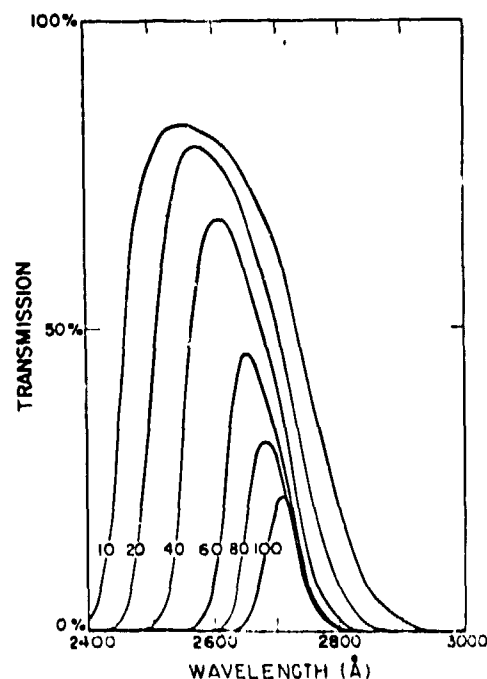
Figure 1. Transmission spectra of 1.0-mm-thick CS_2-hexane mixes. The number associated with each line refers to the percentage (by volume) of CS_2 in the mixture. The transmission increases, broadens, and shifts with CS_2 dilution. These curves have not been corrected for the Fresnel losses of our quartz cell.

Experimental Setup

In our experiments, a single, collimated, 5-mm-diameter, 400 mJ, 15 psec, 2660 Å pulse (quadrupled Nd:YAG) entered the experimental arrangement depicted in Fig. 2. The pulse
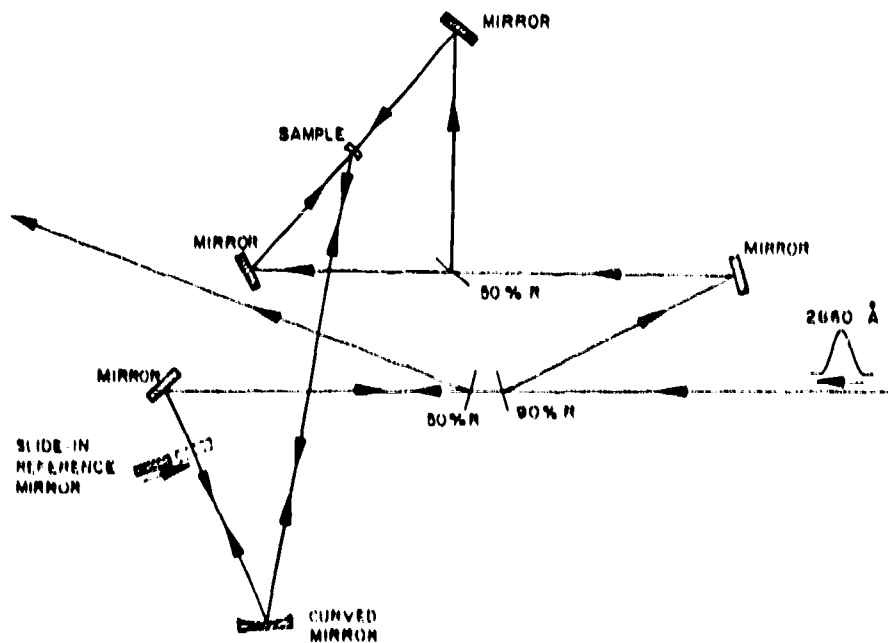

Figure 2. Experimental arrangement. Most of the ultrashort pulse is directed to provide the counterpropagating pump waves; the remainder is brought to the sample as a probe beam.

first encountered a 90% reflecting beamsplitter, and the larger portion was directed into a ring to provide counterpropagating pump waves within the 1-mm sample. The beam which passed the 90% reflecting beamsplitter passed through a 50% reflecting beamsplitter and impinged (as a probe beam) onto the sample at a 30° angle. All beams were linearly polarized in the same direction. A curved mirror focused the probe beam to a point beyond the sample, minimizing the diameter at the sample to insure complete overlap of the three beams. A 00% reflector could be placed before the curved mirror as a reference for evaluating conjugate reflectivities. Any conjugate emission retraced the path of the probe beam, and a portion was redirected by the 50% reflecting beamsplitter for detection and analysis. A uv-sensitive television system (Hamamatsu tube N983) and a video storage device gave continuous two-dimensional display of each conjugate signal's spatial profile, and nanosecond resolution uv-sensitive photomultipliers provided waveforms. A uv-sensitive streak camera measured the various pulse arrival times enabling adjustment of the various optical paths within 3-mm tolerances.

Experimental Results

With a CS_2-hexane solution (60% CS_2 by volume) in a 1-mm cell, signals were observed on the video monitor. Their phase-conjugate nature was confirmed by their disappearance when blocking either of the two pump beams or the probe beam (blocked between the 50% and 90% beamsplitters). Also, the spot on the video monitor did not move with minor angular adjustment of the curved mirror, thereby satisfying a one-dimensional aberration test.

Separate photomultipliers monitored the conjugate and the input signals. The 1.5 nanoseconds response photomultipliers acted as high-speed calorimeters. The conjugate channel gave two peaks; the true conjugate signal was preceeded (by 3 ns) by scattering from the 50% beamsplitter. We measured only the height of the second peak. The data are shown in Fig. 3. We least-squares fit the data to the equation

$$E_s = a \, E_{in} + b(E_{in})^c, \qquad (1)$$

where E_s is the observed signal energy and E_{in} is the input signal energy. The first term represents the residual linear scattering. We found $a = 0.15$, $b = 0.028$, and $c = 2.95$. The value of c is in excellent agreement with the expected value of 3, indicative of the degenerate four-wave mixing process.[5]
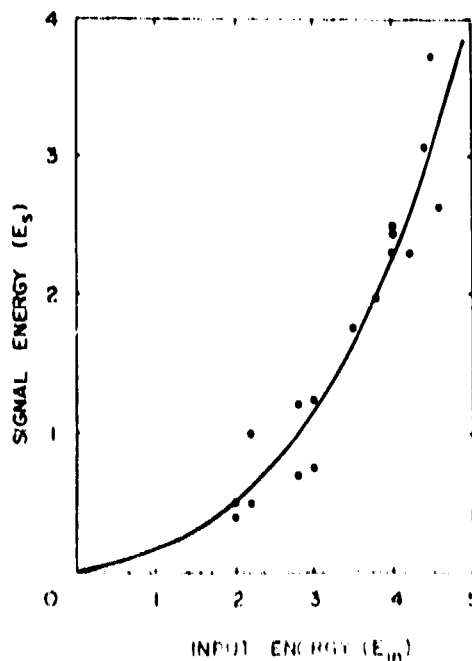

Figure 3. Input versus output energy (arbitrary units) for a 1-mm thick conjugator containing (by volume) 60% CS_2 and 40% hexane. To include spurious scattering, we fit the data to the expression of Equation (1), giving $c = 2.96$, and a nearly cubic dependence for the nonlinear term. The best-fit curve is also drawn.

At the highest input intensities, we obtained 0.1% conjugate reflectivity. To compare these results to theory, the simple model of degenerate four-wave mixing must be modified to include nonsaturable absorption. The low-reflectivity expression becomes:

$$R = [k\ell\delta n_o \exp(-\alpha\ell)]^2, \qquad (2)$$

where $k = \omega/c$, δn_o is the nonlinear index change induced by one pump wave at its entry face, and α is the nonsaturable absorption coefficient. To estimate the induced index change from the pump, we note that $\delta n = \frac{1}{2}n_2\mathscr{E}^2$, where $\mathscr{E}$ is the slowly varying envelope function of one of the pumps. For CS_2, $n_2 \sim 4.34 \times 10^{-11}$ esu at 2660 Å.[1,6] We assume for a 60% CS_2 mix that n_2 is reduced by a factor 0.6 (ignoring small local field and volumetric corrections[6]). The peak intensity of each pump was 40 MW/cm^2 corresponding to $\delta n_o = 3.1 \times 10^{-6}$. Through Eq. (2), this gives R = 0.12%, in good agreement with the measured value.

The signal vanishes for CS_2 concentrations below 5%. Furthermore, introducing a 40 psec delay into either of the two pump paths decreased the 60% CS_2-generated conjugation signal by more than one order of magnitude. The signal disappearance with dilution and reduction with delay rule out the roles of solvated electrons[7] and thermal gradients,[8] respectively, leaving only the optical Kerr effect in CS_2 as the dominant nonlinearity responsible for our conjugate signals.

Phase conjugate signals of approximately the same reflectivity were obtained for 60% by volume CS_2 mixtures in cyclohexane, ethyl alcohol, 1-2 dichloroethane, and N-butanol. These results indicate that the n_2 of dilute CS_2 is note strongly solvent-dependent. We have also seen weaker phase conjugation in pure DMSO, in 1,2 dichloroethane, and ß-carotene-hexane mixes. Because of the high absorption, no signals were seen in pure CS_2.

In addition to the above Kerr-conjugators, we have studied conjugation in various saturable absorbers. At present, the precise mechanism for the observed effects is not clear. Heating,[8] saturation,[9] and photochemical reactions[10] may play roles. Florescein in ethyl alcohol, rhodamine 6G in water or in ethyl alcohol, Kodak A9740 dye in 1,2 dichloroethane, pyrene in hexane, and 2-Napthol 6-Sulfonate ir water (pH 4.5) all exhibited 0.01% conjugate reflectivity. Neither water nor ethanol gave a signal.

Conclusion

In conclusion, we have obtained the first demonstration of phase conjugation in the uv. Because we used ultrashort pulses, sample thicknesses were limited to millimeter dimensions. For lasers with greater coherence lengths, thicker samples could be used with correspondingly higher conjugate reflectivities. Recently, Murray et al.[11] used intra-cavity etalon techniques to reduce the 5 Å free-running KrF laser linewidth to 0.15 cm^{-1}, and Hawkins et al.[12] used KrF amplification of frequency-doubled dye laser pulses to obtain 0.005 cm^{-1} bandwidths. These narrow-band laser systems should allow conjugate reflectivities approaching 100% in thicker cells containing some of the Kerr and saturable absorber media reported here.

Acknowledgments

References

1. J. W. Lewis and W. H. Orttung [J. Phys. Chem. 82, 698 (1978)] indicate that the DC Kerr constant at 2660 Å is seven times that at 5500 Å, and, therefore, n_2 is enhanced by ~ 3.5.
2. R. W. Hellwarth, J. Opt. Soc. Am. 67, 1 (1977); A. Yariv and D. M. Pepper, Opt. Lett. 1, 16 (1977); D. Bloom and G. C. Bjorklund, Appl. Phys. Lett. 31, 592 (1977); A. Yariv, IEEE J. Quantum Electron. QE-14, 650 (1978) and references cited therein.
3. N. F. Pilipetskii, V. I. Popovichev and V. V. Ragul'skii, JETP Lett. 27, 585 (1978); G. V. Peregudov, M. E. Plotkin, and E. N. Ragozin, Sov. J. Quantum. Electron. 9, 1413 (1979); A. A. Ilyukhin, G. V. Peregudov, M. E. Plotkin, E. N. Ragozin, and V. A. Chirkov, JETP Lett. 29, 328 (1980); D. T. Hon, 11th Int. Quant. Electron. Conf. Digest, and JOSA 70, 635 (1980).

4. W. Liptay, Angew. Chem. Internat. Edit. 8, 177 (1969); P. Baraldi, P. Mirone, and E. S. Guidetti, Z. Naturforsch. 26a, 1852 (1971).

5. For a low-reflectivity conjugator, the reflectivity is proportional to the square of the pump wave intensity. The probe signal is also proportional to the pump wave intensity, giving overall cubic dependence. See, for example, E. E. Bergmann, I. J. Bigio, P. J. Feldman and R. A. Fisher, Opt. Lett. 3, 82 (1978).

6. R. W. Hellwarth and N. George, Opto-Electron. 1, 213 (1969).

7. G. A. Kenney-Wallace and C. D. Jonah, Chem. Phys. Lett. 47, 362 (1977).

8. G. Martin and R. W. Hellwarth, Appl. Phys. Lett. 34, 371 (1979).

9. R. L. Abrams and R. C. Lind, Opt. Lett. 2, 94 (1978), and Erratum, Opt. Lett. 3, 205 (1978).

10. J. H. Clark, S. L. Shapiro, A. J. Campillo, and K. R. Winn, J. Am. Chem. Soc. 101, 747 (1979).

11. J. R. Murray, J. Goldhar, D. Eimerl and A. Szoke, IEEE J. Quant. Electron. QE-15, 342 (1979).

12. R. T. Hawkins, H. Egger, J. Bokor and C. K. Rhodes, Appl. Phys. Lett. 36, 391 (1980).